

# Phase Separation Kinetics in a Model with Order-Parameter Dependent Mobility

by

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## Abstract

We present extensive results from 2-dimensional simulations of phase separation kinetics in a model with order-parameter dependent mobility. We find that the time-dependent structure factor exhibits dynamical scaling and the scaling function is numerically indistinguishable from that for the Cahn-Hilliard (CH) equation, even in the limit where surface diffusion is the mechanism for domain growth. This supports the view that the scaling form of the structure factor is "universal" and leads us to question the conventional wisdom that an accurate representation of the scaled structure factor for the CH equation can only be obtained from a theory which correctly models bulk diffusion.

# 1 Introduction

When a two-phase mixture in a homogeneous phase is quenched below the critical coexistence temperature, it becomes thermodynamically unstable and evolves towards a new equilibrium state, consisting of regions which are rich in one or the other constituent of the mixture. The dynamics of this evolution is referred to as "phase ordering dynamics" and constitutes a well-studied problem in nonequilibrium statistical mechanics [1]. As a result of these investigations, there is now a good understanding of many aspects of phase ordering in pure and isotropic binary mixtures. Thus, it is generally accepted that the coarsening domains are characterised by a unique, time-dependent length scale  $L(t)$ , where  $t$  is time. Furthermore, the nature of the phase ordering process depends critically on whether or not the order parameter is conserved. For systems characterised by a nonconserved order parameter, e.g., ordering of a ferromagnet, growing domains obey the Lifshitz-Cahn-Allen (LCA) growth law  $L(t) \sim t^{\frac{1}{2}}$  [1]. For systems with a conserved order parameter but no hydrodynamic effects, e.g., segregation of a binary alloy, the characteristic domain size obeys the Lifshitz-Slyozov (LS) growth law  $L(t) \sim t^{\frac{1}{3}}$  [1]. For systems with a conserved order parameter and hydrodynamic effects, e.g., segregation of a binary liquid, there appear to be various regimes of domain growth, depending on the dimensionality and system parameters [2, 3].

As far as the analytic situation is concerned, there is a reasonable understanding of the nonconserved case for pure and isotropic systems. In particular, the LCA diffusive growth law has been derived in some exact models [4]. In addition, Ohta et al. and Oono and Puri [5] have proposed an analytic form for the time-dependent structure factor which is in good agreement with numerical results, though the quality of this agreement has recently been questioned by Blundell et al. [5]. For the conserved case, the situation is less satisfactory. There is some understanding of the growth exponents and one has a good empirical form for the scaled structure factor – at least without hydrodynamics [7]. However, this functional form is analytically derivable only in the limiting case where one of the components is present in a small fraction [1]. An outstanding theoretical problem in this field is the calculation of the scaled structure factor for the conserved case when the two components of the mixture are present in an equal proportion, viz., the so-called critical quench [8]. Our results in this paper provide some interesting insights on this problem, as we will discuss later.

In this paper, we study a model for phase separation dynamics in systems where the mobility is order-parameter dependent. We will present detailed numerical results from a simulation of this model. This paper is organised as follows. In Section 2, we briefly discuss our model and its static solution. In Section 3, we present numerical results obtained from our model. Section

4 ends this paper with a summary and discussion.

## 2 Model for Phase-Separating Systems with Order-Parameter Dependent Mobility

The dynamics of phase separation is usually described by the phenomenological equation

$$\frac{\partial\phi(\vec{r},t)}{\partial t} = \vec{\nabla} \cdot \left[ M(\phi) \vec{\nabla} \left( \frac{\delta H[\phi(\vec{r},t)]}{\delta\phi(\vec{r},t)} \right) \right], \quad (1)$$

where  $\phi(\vec{r},t)$  is the order parameter at point  $\vec{r}$  and time  $t$  and is a measure of the local difference in densities of the two segregating species, say A and B. In (1),  $M(\phi)$  corresponds to the mobility, which is dependent on the order parameter, in general. The free-energy functional is usually chosen to be of the standard  $\phi^4$ -form, viz.,

$$H[\phi(\vec{r},t)] = \int d\vec{r} \left[ -\frac{1}{2}\phi(\vec{r},t)^2 + \frac{1}{4}\phi(\vec{r},t)^4 + \frac{1}{2}(\vec{\nabla}\phi(\vec{r},t))^2 \right], \quad (2)$$

where we assume that all variables have been rescaled into dimensionless units; and the system is below the critical temperature. The dynamics of Eqs. (1)-(2) drives the order parameter to the local fixed point values  $\phi_0 = \pm 1$ , corresponding to (say) A- and B-rich phases, respectively. The temporal evolution described by Eq. (1) also satisfies the conservation constraint that  $\int d\vec{r}\phi(\vec{r},t)$  is constant in time.

There have been many studies of Eq. (1) in the limiting case of the Cahn-Hilliard (CH) equation [9], where the mobility is constant, viz.,  $M(\phi) = 1$  (in

dimensionless units). Numerical studies of the CH equation and equivalent Cell Dynamical System (CDS) models [10] demonstrate that late-stage domain growth obeys the LS growth law we have quoted earlier (i.e.,  $L(t) \sim t^{\frac{1}{3}}$ ). These studies also clarify the functional form of the scaled structure factor which characterises the morphology of the coarsening domains.

For deep quenches, it has been pointed out by Langer et al. and Kitahara and Imada [11] that a more realistic model for phase separation should explicitly incorporate an order-parameter dependent mobility of the form

$$M(\phi) = 1 - \alpha\phi^2, \quad (3)$$

where  $\alpha$  parametrises the depth of the quench. At the physical level, this form of the mobility can be understood as follows. Deep quenches result in enhanced segregation in that A-rich (or B-rich) domains are purer in A (or B) than in the case of shallow quenches. Thus, if one presumes that phase separation occurs by exchanges of neighbouring A- and B-atoms, the probability of such an exchange in the bulk is drastically reduced for deep quenches. This can be mimicked by the order-parameter dependent mobility in (3) with  $\alpha \rightarrow 1$ . At the mathematical level, Kitahara and Imada [11] have shown that an order-parameter dependent mobility arises naturally if one attempts to obtain a coarse-grained model for phase separation from a master equation description of an appropriate microscopic model, viz., the Ising model with Kawasaki spin-exchange kinetics [12].

The physical effect of the order-parameter dependent mobility is that, as  $\alpha \rightarrow 1$  (which happens for temperature  $T \rightarrow 0$ ), bulk diffusion is substantially suppressed because the mobility  $M(\phi_0) \rightarrow 0$ . Therefore, the effects of surface diffusion are relatively enhanced. The surface-diffusion mechanism for domain growth has an associated growth law  $L(t) \sim t^{\frac{1}{4}}$  [13], in contrast to the evaporation-condensation mechanism which drives asymptotic growth in the CH equation and gives rise to the LS growth law. Therefore, as  $T \rightarrow 0$ , one expects an extended regime of  $t^{\frac{1}{4}}$  growth in the dynamics of Eqs. (1)-(3). This model has been studied numerically by various authors [14] and we will remark on their results shortly. Furthermore, Bray and Emmott [15] have analytically studied phase separation in models with order-parameter dependent mobility in the limit where one of the components is present in a vanishingly small fraction. In passing, we should also point out that an order-parameter dependent mobility as in (3) has proven to be a useful way of incorporating the effects of external fields which vary linearly with distance, e.g., gravity. However, we will not go into this here and merely refer the interested reader to Reference [16].

In recent work, there was proposed a novel dynamical equation for phase separation in binary mixtures – using the master equation formulation for an Ising model with Kawasaki spin-exchange kinetics [17]. This equation was first obtained in the context of phase separation in a gravitational field but

does not reduce to the CH equation in the absence of gravity. As a matter of fact, it takes a form similar to that of Eq. (1), i.e.,

$$\frac{\partial\phi(\vec{r},t)}{\partial t} = \vec{\nabla} \cdot \left[ (1 - \phi(\vec{r},t)^2) \vec{\nabla} \left( \frac{\delta H[\phi(\vec{r},t)]}{\delta\phi(\vec{r},t)} \right) \right], \quad (4)$$

with the free energy

$$\begin{aligned} H[\phi(\vec{r},t)] = & \frac{T}{T_c - T} \int d\vec{r} \frac{1}{2} \left[ (1 + \phi(\vec{r},t)) \ln(1 + \phi(\vec{r},t)) \right. \\ & + (1 - \phi(\vec{r},t)) \ln(1 - \phi(\vec{r},t)) - \frac{T_c}{T} \phi(\vec{r},t)^2 \\ & \left. + \frac{T_c - T}{T} (\vec{\nabla}\phi(\vec{r},t))^2 \right]. \end{aligned} \quad (5)$$

Eqs. (4)-(5) have been cast in a dimensionless form by a rescaling of the space and time variables analogous to that for the CH equation [17]. (Clearly, this rescaling is not appropriate in the vicinity of the critical temperature  $T_c$ .) It is difficult to put Eqs. (4)-(5) in a parameter-free form because of the additional term in comparison to the CH equation and the nature of the static solution, which we discuss below. The first two terms under the integral sign in (5) are recognised as the entropy of a noninteracting binary mixture and the next two terms correspond to the interaction part [18].

Eqs. (4)-(5) have the pleasant feature that they explicitly contain the mean-field static solution  $\phi^s(\vec{r})$ , which is the solution of

$$\phi^s(\vec{r}) = \tanh \left[ \frac{T_c}{T} \phi^s(\vec{r}) + \left( \frac{T_c}{T} - 1 \right) \nabla^2 \phi^s(\vec{r}) \right], \quad (6)$$

where it should be kept in mind that the space variable has been rescaled. However, we do not expect our model to be in a different dynamical univer-

sality class from Eqs. (1)-(3). In our model, as  $T \rightarrow 0$ , the saturation value of the order parameter  $\phi_0 \rightarrow \pm 1$ . This reduces the bulk diffusion because of the order-parameter dependent mobility and enhances the time-regime in which one observes surface-diffusion mediated growth. In the case where surface diffusion is predominant, we follow the terminology established by Hohenberg and Halperin [19] and refer to our model as "Model S", where S refers to surface diffusion. In the classification of Hohenberg and Halperin, the CH equation is referred to as Model B. For shallow quenches, the saturation value of the order parameter  $\phi_0$  is considerably less than 1 and the mobility  $M(\phi)(= 1 - \phi^2)$  is not significantly reduced in the bulk. In this limit, the dynamics of our model is in the same dynamical universality class as Model B or the CH equation.

In this paper, we present detailed numerical results from a simulation of (4)-(5). The purpose of this paper is two-fold. Firstly, our numerical results improve substantially upon existent results [14] for models with order-parameter dependent mobility. Secondly, we believe that our results may be of some relevance to an outstanding theoretical problem of phase separation dynamics, viz., the computation of the scaling form of the time-dependent structure factor.

Before we present numerical results, we would like to briefly discuss the interfacial profile in our model. For this, we need the solution of the 1-

dimensional version of (6), viz.,

$$\frac{d^2\phi^s(x)}{dx^2} = -\frac{T_c}{T_c - T}\phi^s(x) + \frac{T}{T_c - T}\tanh^{-1}(\phi^s(x)). \quad (7)$$

Multiplying both sides by  $2(d\phi^s(x)/dx)$ , we can trivially integrate this equation to get

$$\begin{aligned} \frac{d\phi^s(x)}{dx} &= \left[ \frac{2T}{T_c - T}\phi^s(x)\tanh^{-1}(\phi^s(x)) + \frac{T}{T_c - T}\ln\left(\frac{1 - \phi^s(x)^2}{1 - \phi_0^2}\right) - \right. \\ &\quad \left. \frac{T_c}{T_c - T}(\phi^s(x)^2 + \phi_0^2) \right]^{\frac{1}{2}}, \end{aligned} \quad (8)$$

where we focus on the profile which goes from  $-\phi_0$  at  $x = -\infty$  to  $\phi_0$  at  $x = \infty$ . A second integration is only possible numerically and we show the resultant profiles for  $x > 0$  in Figure 1(a) for four different values of  $T/T_c$ . This solution has the form  $\phi^s(x) = \phi_0 f(x/\xi)$ , where  $f(y)$  is a sigmoidal function and  $\xi$  measures the correlation length or interface thickness in dimensionless units. An estimate of  $\xi$  is obtained as the distance over which  $f(x/\xi)$  rises from 0 to (say)  $1/\sqrt{2}$  of its maximum value. The profiles as a function of the scaled distance  $x/\xi$  are shown in Figure 1(b). They do not exhibit a universal collapse because of a weak dependence of  $f(y)$  on the parameter  $T/T_c$ . In any case, our interest in the correlation length is primarily from a numerical standpoint in that the discretisation mesh size in space should not exceed the interface thickness, which is approximately  $2\xi$ .

### 3 Numerical results

We have conducted extensive 2-dimensional numerical simulations of (4)-(5) for the parameter values  $T/T_c = 0.2, 0.4, 0.5$  and  $0.8$ , corresponding to  $\phi_0 \simeq 0.9999, 0.9857, 0.9575$  and  $0.7105$ , respectively. We implement a simple Euler discretisation of (4)-(5) on a lattice of size  $N \times N$ . The Laplacian and divergence operators in (4)-(5) are replaced by their isotropically discretised equivalents, involving both nearest and next-nearest neighbours. The discrete implementation of our model with order-parameter dependent mobility has the unpleasant feature that it is unstable for  $\phi > 1$  and numerical fluctuations which cause  $\phi$  to become larger than 1 give rise to unphysical divergences. (This property is common to all such models [14].) For  $T/T_c = 0.2 (\phi_0 \simeq 0.9999)$ , this causes a numerical problem because of the proximity of the saturation value to  $\pm 1$ . We circumvent this problem by using a very fine mesh size ( $\Delta t = 0.001$  and  $\Delta x = 0.5$ ) and by setting the value of  $\phi$  equal to  $\phi_0$  (or  $-\phi_0$ ) whenever it exceeds  $\phi_0$  (or becomes less than  $-\phi_0$ ). We have confirmed that this procedure does not cause any appreciable violation of order parameter conservation for the extremely fine mesh we have used. For the higher values of  $T$  studied here, we use the coarser mesh sizes  $\Delta t = 0.01$  and  $\Delta x = 1.0$  and this suffices for our purposes.

Periodic boundary conditions are applied in both directions of our lattice. For all simulations described here, the initial condition for the order

parameter consists of a uniformly distributed random fluctuation of amplitude 0.025 about a zero background. This mimics a critical quench from high temperatures, at which the system is homogeneous but has small thermal fluctuations.

Apart from evolution pictures and profiles, the statistical quantity of experimental interest is the time-dependent structure factor

$$S(\vec{k}, t) = \langle \phi(\vec{k}, t) \phi(\vec{k}, t)^* \rangle, \quad (9)$$

which is the Fourier transform at wave-vector  $\vec{k}$  of the order parameter correlation function. In (9),  $\phi(\vec{k}, t)$  is the Fourier transform of  $\phi(\vec{r}, t)$  and the angular brackets refer to an averaging over an ensemble of initial conditions. In our discrete simulations, the wave-vector  $\vec{k}$  takes the discrete values  $\frac{2\pi}{N\Delta x}(n_x, n_y)$ , where  $n_x$  and  $n_y$  range from  $-N/2$  to  $(N/2) - 1$ . We present here structure factor data obtained on  $512 \times 512$  systems as an average over 60 independent initial conditions. The order parameter profiles are hardened before computing the structure factor, viz., the values of  $\phi > 0$  are set equal to 1 and  $\phi < 0$  are set equal to -1. The structure factor is normalised as  $\sum_{\vec{k}} S(\vec{k}, t)/N^2 = 1$ . All results presented below are for the spherically averaged structure factor  $S(k, t)$ .

Experimentalists are typically interested in whether or not the structure factor exhibits dynamical scaling [20], viz., whether or not the time-dependence of the spherically averaged structure factor has the simple scaling

form

$$S(k, t) = L(t)^d F(kL(t)), \quad (10)$$

where  $d$  is the dimensionality and  $F(x)$  is a time-independent master function. The interpretation of dynamical scaling is that the coarsening pattern maintains its morphology but the characteristic length scale  $L(t)$  increases with time. There are many equivalent definitions (upto prefactors) of the characteristic length scale. We use what is perhaps the most commonly-used definition, viz., the inverse of the first moment of the spherically averaged structure factor  $S(k, t)$ . Thus, we have  $L(t) = \langle k \rangle^{-1}$ , where

$$\langle k \rangle = \frac{\int_0^{k_m} dk k S(k, t)}{\int_0^{k_m} dk S(k, t)}. \quad (11)$$

In (11), we take the upper cut-off  $k_m$  as half the magnitude of the largest wavevector in the Brillouin zone. At these large values of the wavevector, the structure factor has decayed to approximately zero and the value of  $\langle k \rangle$  is unchanged even if we increase the cutoff. Of course, one could also define a length scale using higher moments of the structure factor or zeroes of the correlation function. However, in the dynamical scaling regime [20], these definitions are all equivalent.

Figure 2 shows evolution pictures from a disordered initial condition for the parameter value  $T/T_c = 0.2$  (or  $\phi_0 \simeq 0.9999$ ) and a lattice size  $256 \times 256$ . This low value of temperature corresponds to a situation in which there is almost no bulk diffusion once the order parameter saturates out to its

equilibrium values. In this case, domain growth occurs via surface diffusion and has an associated growth law  $L(t) \sim t^{\frac{1}{4}}$  [13]. Notice that the domain morphology in this case is considerably different from the morphology in the usual CH case with the bicontinuous domains being more serpentine and intertwined in the present case. Figure 3 shows the corresponding evolution pictures from a  $256 \times 256$  lattice for  $T/T_c = 0.5$  (or  $\phi_0 \simeq 0.9575$ ). These pictures are more reminiscent of the CH morphology. Figure 4 shows the variation of order parameter along a horizontal cross-section at the middle of the lattice for the evolution pictures of Figure 2. Figure 5 shows the order parameter profiles corresponding to the evolution depicted in Figure 3. These profiles provide a qualitative measure of the thinning out of defects (viz., interfaces) as the coarsening proceeds.

In Figure 6(a), we superpose data from different times for the scaled structure factor  $S(k, t)\langle k \rangle^2$  vs.  $k/\langle k \rangle$ . The parameter value is  $T/T_c = 0.2$ , corresponding to growth mediated by surface diffusion (i.e., Model S). The structure factor data collapses neatly onto a master curve, exhibiting the validity of dynamical scaling in this system. The solid line refers to the scaled structure factor for the CH equation obtained with the same system sizes and statistics as described previously. On the scale of this figure, the scaled structure factor for Model S is coincident with that for the CH equation except for the first two points after  $k = 0$ , which exhibit violation of scaling

because of finite-size effects. A similar observation has also been made for the real-space correlation function by Lacasta et al. [14]. However, we should stress that the structure factor is a more sensitive characteristic of phase ordering dynamics than the correlation function. Furthermore, our present data (obtained on  $512 \times 512$  systems with 60 independent runs and  $\Delta t = 0.001$ ,  $\Delta x = 0.5$ ) constitutes a considerable improvement over that of Lacasta et al. [14], who used a  $120 \times 120$  system with 10 independent runs and  $\Delta t = 0.025$ ,  $\Delta x = 1.0$ .

Before we proceed, two further remarks are in order. Firstly, it is interesting that the structure factors for Model S and the CH model are numerically indistinguishable, even though the morphologies are different and domain growth is characterised by different power laws. Clearly, the time-dependent structure factor (which is the Fourier transform of the equal-time correlation function) is not a sufficiently good measure of the morphology to discriminate between these two situations and perhaps one needs to invoke other tools like two-time correlation functions or higher-order structure factors [6]. Nevertheless, the structure factor is an experimentally relevant quantity and the computation of its analytic form for the CH equation has been an outstanding problem to date. Furthermore, it has been believed that a "correct" theory for the scaling form of the structure factor must properly account for the bulk diffusion and the LS growth law [8, 21]. However, our numerical

results demonstrate that the scaling form of the structure factor for the conserved case is considerably robust and is not affected by the growth exponent or the underlying growth mechanism, at least for the model we have studied.

The second remark we wish to make concerns the dashed line in Figure 6(a), which is obtained from a naive application of the theory of Mazenko [21], who developed a Gaussian closure for the CH equation. The naive Mazenko theory predicts that the asymptotic growth law is  $L(t) \sim t^{\frac{1}{4}}$  rather than the numerically observed LS law, viz.,  $L(t) \sim t^{\frac{1}{3}}$ . Because of the lower growth exponent, it is presumed that the naive Mazenko theory describes the surface-diffusion growth regime of the CH equation. In the light of our present results, it is clear that the form of the scaled structure factor is largely independent of the mechanism of domain growth. Unfortunately, as is clear from Figure 6(a), the analytic form obtained from the naive Mazenko theory is not correct in most respects and only gets right the approximate width of the scaling function. We are presently investigating a Gaussian closure of (4) to see whether it gives better results for the scaling function.

Figure 6(b) plots the data of Figure 6(a) on a log-log scale and reconfirms the coincidence of the CH and Model S scaling functions, including the Porod tail  $S(k, t) \sim k^{-3}$  for large  $k$ . At small values of  $k$ , the scaled structure factor for Model S exhibits a  $k^4$ -behaviour as in the CH case [22], except for the first couple of values of  $k$ , which are probably affected by finite-size effects.

Again, the dashed line is from the naive Mazenko theory and has the wrong behaviour for small values of  $k$ , viz.,  $S(k, t) \sim k^2$  rather than  $S(k, t) \sim k^4$ . The analytic form matches the numerical results in the Porod tail but this may be entirely fortuitous. Figure 6(c) plots the data of Figure 6(a) on a Porod plot, viz.,  $k^4 S(k, t)/\langle k \rangle^2$  vs.  $k/\langle k \rangle$ , which highlights features of the Porod tail. In this case, our data is not reliable for  $k/\langle k \rangle \geq 2.5$ . However, upto that point, the scaled form factors for the Model S and CH cases are again indistinguishable, including the first valley after the peak [8].

Similar results for the scaled structure factor are found for higher values of temperature  $T$  also. This is not surprising as the morphology for our model goes over to that for the CH equation at higher values of the temperature (see Figure 3). For the sake of brevity, we do not show structure factor data for higher values of  $T$ .

Figure 7(a) shows the time-dependent length scale  $L(t)$  as a function of dimensionless time  $t$  for four different values of temperature ( $T/T_c = 0.2, 0.4, 0.5$  and  $0.8$ ) in our model. Recall that surface diffusion effects are enhanced as  $T$  is lowered because  $\phi_0 \rightarrow 1$  as  $T \rightarrow 0$ . For purposes of comparison, we have also included the length scale data for the CH equation. Figure 7(b) is a log-log plot of the data in Figure 7(a). We use a fitting routine to fit a straight line to the data. The resultant exponents (denoted as  $x$ ) for the CH equation and the case with  $T/T_c = 0.8$  are identical, viz.,  $x = 0.33$ .

On the other hand, for  $T/T_c = 0.2$ , we again get a straight line but the associated growth exponent is 0.25, which is associated with domain growth via surface diffusion [13, 14]. For intermediate values of  $T/T_c$  (viz., 0.4 and 0.5), we do not get a good linear fit as the length scale is in a transition regime between  $L(t) \sim t^{\frac{1}{4}}$  and  $L(t) \sim t^{\frac{1}{3}}$ .

## 4 Summary and Discussion

Let us end this paper with a brief summary and discussion of our results. We have presented detailed results from an extensive numerical simulation of a model with order-parameter dependent mobility. We expect this model to be in the same dynamical universality class as other models with order-parameter dependent mobility [11, 14] but it has the additional pleasant feature that it explicitly contains the mean-field static solution.

Because of the large system sizes and extensive averaging employed by us, we are able to obtain the best numerical results on such systems to date. The salient features of our results are as follows. In the parameter regime where surface diffusion drives domain growth, the morphology of evolving patterns is more serpentine than that in the CH equation. However, the scaling form of the time-dependent structure factor for surface-diffusion mediated growth appears to be numerically identical to that for the CH equation, including the Porod tail and the small- $k$  behaviour. This numerical result casts doubts

on the conventional wisdom that a "correct" theory for the scaling form of the CH structure factor must contain the correct growth law and properly model the bulk diffusion field. As a matter of fact, we are led to speculate that the scaling form for the conserved case may be dictated by more general considerations, e.g., domain-size distributions, etc. This is an approach we are presently pursuing in an attempt to obtain a better understanding of the functional form of the structure factor for the conserved case.

We are also interested in examining other models of phase separation to see whether they give rise to similar results for the scaled structure factor. In particular, Giacomin and Lebowitz [23] have recently studied an Ising model on a cubic lattice with Kawasaki spin-exchange kinetics which satisfies detailed balance. The spins interact via a long-ranged Kac interaction potential of the form  $V(r_{ij}) = \gamma^d J(\gamma r_{ij})$ , where  $r_{ij}$  is the distance between spins  $i$  and  $j$ ;  $\gamma$  is a parameter; and  $d$  is the dimensionality. In the limit  $\gamma \rightarrow 0$ , Giacomin and Lebowitz rigorously obtain an exact nonlinear evolution equation for phase separation. Their model is of the same form as Eqs. (4)-(5) but contains a nonlocal interaction term, instead of the gradient square term in (5). They argue that this exact equation gives results for interface motion which are similar to those obtained from the CH equation. We are interested in examining whether or not this exact equation is in the same dynamical universality class as the CH equation.

Finally, we should point out that the difference in morphologies between Model S and the CH equation must show up at some level, e.g., two-time correlation functions or higher-order structure factors [6]. This is another question we are presently interested in. Nevertheless, this possible difference in two-time correlation functions or higher-order structure factors does not detract from the relevance of the fact that the scaled form of the conventional structure factor is very robust. After all, the conventional structure factor is the primary quantity of experimental, numerical and theoretical interest.

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## Figure Captions

Figure 1 : (a) Static wall solutions of the model described in the text (Eqs. (4)-(5)). The solutions are obtained by numerically solving (8). We plot the profile  $\phi^s(x)/\phi_0$  vs.  $x$  for  $x > 0$  (where  $\phi_0$  is the saturation value) for four values of the temperature  $T$ , viz.,  $T/T_c = 0.2, 0.4, 0.5$  and  $0.8$ .  
(b) Same as (a) except the distance  $x$  is scaled by a correlation length  $\xi$ , which is defined as the distance over which the wall profile rises to  $1/\sqrt{2}$  of its maximum value.

Figure 2 : Evolution pictures from a disordered initial condition for an Euler-discretised version of (4)-(5) on a  $256 \times 256$  lattice. Regions with positive order parameter are marked in black and those with negative order parameter are not marked. The parameter value is  $T/T_c = 0.2$ , corresponding to a situation in which surface diffusion is the primary mechanism of domain growth. The discretisation mesh sizes are  $\Delta t = 0.001$  and  $\Delta x = 0.5$ . Periodic boundary conditions are applied in both directions. The initial condition consists of uniformly-distributed random fluctuations of amplitude 0.025 about a zero background, corresponding to a critical quench. The evolution pictures are shown for dimensionless times 1000, 2000, 4000 and 10000.

Figure 3 : Similar to Figure 2 but for the parameter value  $T/T_c = 0.5$ .

Figure 4 : Order parameter profiles for the evolution depicted in Figure 2. The profiles are measured along a horizontal cross-section at the centre of the vertical axis.

Figure 5 : Order parameter profiles for the evolution depicted in Figure 3. The cross-section is the same as that for Figure 4.

Figure 6 : (a) Superposition of scaled structure factor data from a simulation of (4)-(5) with  $T/T_c = 0.2$ , corresponding to the surface-diffusion case. We plot  $S(k, t)\langle k \rangle^2$  vs.  $k/\langle k \rangle$  for data from dimensionless times 2000, 3000, 4000 and 10000. The structure factor is computed on a  $512 \times 512$  lattice as an average over 60 independent initial conditions. It is normalised as described in the text and then spherically averaged. The first moment of  $S(k, t)$  is denoted as  $\langle k \rangle$  and measures the inverse of the characteristic length scale. The solid line is a scaled plot of structure factor data from the CH equation at dimensionless time 10000. Finally, the dashed line is an analytic form obtained from a naive application of Mazenko theory [21], which yields the domain growth law  $L(t) \sim t^{\frac{1}{4}}$ .  
(b) Plot of data from (a) on a log-log scale. The Porod tail is extracted by hardening the order parameter field before computing the structure factor.  
(c) Porod plot (viz.,  $k^4 S(k, t)/\langle k \rangle^2$  vs.  $k/\langle k \rangle$ ) for the data from (a). This plot highlights the features of the Porod tail. Unfortunately, our

data in this plot exhibits large fluctuations for  $k/\langle k \rangle \geq 2.5$ .

Figure 7 : (a) Characteristic domain size  $L(t)$  plotted as a function of dimensionless time for our model in (4)-(5) with  $T/T_c = 0.2, 0.4, 0.5$  and  $0.8$ . For comparison, we also present length scale data from a simulation of the CH equation. The length scale is obtained as the inverse of the first moment of the structure factor  $\langle k \rangle$ .  
(b) Data from (a), plotted on a log-log scale. We use a fitting routine to fit a linear function to the length scale data. The resultant fit (wherever reasonable) is shown on the appropriate data set as a solid line and the corresponding exponent (denoted as  $x$ ) is specified on the figure.